

The University of Chicago

ATOMIC DISINTEGRATION AS OBSERVED IN THE WILSON CHAMBER

A DISSERTATION SUBMITTED TO THE
GRADUATE FACULTY IN CANDIDACY FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY

DEPARTMENT OF CHEMISTRY

1930

By
WILLIAM JOSEPH ARNER

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Introduction

The first evidence of atomic disintegration by bombardment with alpha particles was obtained by Rutherford¹. In a series of experiments he and Chadwick² succeeded in disintegrating boron, nitrogen, fluorine, sodium, aluminium, and phosphorus. For the detection of the disintegrations they used what is known as the "scintillation method," and thus obtained evidence of atomic particles of a greater range than the original alpha particles. In the case of nitrogen particularly, they proved that the mass and charge of these atomic particles is identical with that of a hydrogen nucleus or proton. A similar proof was given for three of the other elements. In addition, atomic particles were obtained from all of the elements from boron to potassium inclusive, with the exception of carbon and oxygen. But for these other elements the range and yield of the particles was much less. That is, while boron, nitrogen, fluorine, sodium, aluminium, and phosphorus were disintegrated fairly readily, the other elements proved to be more stable in agreement with the Harkins Rule.

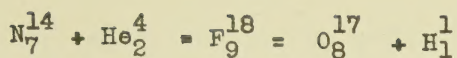
¹E. Rutherford, Phil. Mag., 37, 537, (1919).

²E. Rutherford and J. Chadwick, Phil. Mag., 42, 809 (1921).

In 1923 Harkins and Ryan¹ developed a new method for the determination of what occurs when atoms of nitrogen or other gases are disintegrated by bombardment with alpha particles, and their procedure has shown that what occurs is in a primary sense an atomic synthesis, accompanied by a disintegration. They used the photographic ray track apparatus devised by C.T.R. Wilson,² and modified by Shimizu.³ The fast alpha particles from thorium C' were shot into the chamber from a thorium active deposit. They considered that the tracks of the alpha particles, and of the reaction products produced by the bombardment of atom nuclei, would reveal the mechanism of the atomic disintegration.

The first gas used by them was nitrogen. The method of Harkins and Ryan¹ was adopted without essential change by Blackett,⁴ Harkins and Shadduck,⁵ and Harkins and Schuh.⁶ All of these obtained photographs of the atomic disintegration of nitrogen. The yields obtained were: eight for 270,000, two for 250,000 and two for 250,000 tracks of thorium-C' particles, respectively. An analysis of these pictures revealed that the process is probably:

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- ¹Harkins and Ryan, J. Amer. Chem. Soc. 45, 2095, (1923).
²C.T.R. Wilson, Proc. Roy. Soc., A87, 277, (1912).
³T. Shimizu, Proc. Roy. Soc., A99, 425, (1921).
⁴P.M.S. Blackett, Proc. Roy. Soc., A107, 349, (1925).
⁵Harkins and Shadduck, Proc. Nat. Acad. Sci., 12, 707 (1926).
⁶Harkins and Schuh, Phys. Rev., 35, 809, (1930).



That is, the alpha particle or helium nucleus combines with a nitrogen nucleus to form an isotope of fluorine which instantaneously disintegrates and shoots off both oxygen of mass seventeen and a proton or H-particle of approximately unit mass. In all cases in which the energy relations were tested, there was a lack of conservation of energy in this process, that is the collisions were inelastic.

Kirsch and Pettersson at Vienna used an adaptation of the "scintillation method," and consider that they have found evidence of the disintegration of other elements in addition to those broken up by Rutherford and Chadwick. They consider instability under bombardment by alpha particles to be a much more common property of the elements. For example, they claim¹ to have disintegrated beryllium, carbon, oxygen, and iron. However this work is questioned by Bothe and Franz² who attempted to check their results by the use of a Geiger counter. Chadwick³ in a reconsideration of the earlier work at Cambridge could still find no evidence for the disintegration of any elements in addition to those found in the earlier work. Though there is thus a disagreement

¹G. Kirsch and H. Pettersson, Z. Physik, 42, 641 (1927).

²W. Bothe and H. Franz, Naturwiss., 16, 204 (1928).

³J. Chadwick, Phil. Mag. (7) 2, 1056, (1926).

between the two groups as to the yields and ranges of the H-particles obtained, and the minimum amount of energy of the alpha particles necessary to produce a disintegration, both agree that the rule of Harkins is valid. That is, the odd numbered light elements (boron, nitrogen, fluorine, sodium, aluminium, and phosphorus) are the more readily disintegrated and yield the longest range H-particles.

It would be desirable to obtain more information on the disintegration of these elements, by the method of Harkins and Ryan. This method as developed up to the present is however rather restricted in scope. For greatest suitability the substance examined should be gaseous under ordinary conditions of pressure and temperature; the number of atoms in its molecules should be small, so that the expansion ratio necessary to produce good tracks may not be too large; and it should be comparatively stable in an atmosphere of water vapor. Also, atoms in the molecule other than the one under investigation, should be incapable of disintegration by the alpha particles.

A consideration of compounds of these odd numbered light elements shows them to be generally unsuitable. The hydrides of boron are unsuitable because of their great chemical instability. Boron trichloride might be satisfactory in an atmosphere of carbon tetrachloride. With carbon tetrachloride as the cloud vehicle in place of water, Philipp¹

¹K. Philipp, Z. Physik, 53, 100 (1929).

reports that the rather high expansion ratio of 1.89 is necessary to produce good tracks. The use of such high expansion ratios makes it difficult to prevent surges in the chamber. In addition, the large percentage of chlorine atoms present (three to one of boron plus those from the cloud) would make it practically impossible to determine in any case whether boron or chlorine had been disintegrated. Fluorine and hydrogen fluoride are unsuitable because they act chemically on glass. There are no suitable sodium or aluminium compounds. Phosphorus in phosphine offers the most likely prospect.

If solids can be used, many more elements could be investigated. It was considered, for example, that it might be possible to obtain a complete dynamical picture of the disintegrative process, by placing very thin foils of the substance examined directly in the chamber. Or, a thin layer of the substance to be examined might be spread on a foil of non-disintegrable metal and placed in the chamber in the path of the alpha rays. From this viewpoint, a thin foil of aluminium is the most suitable substance for the purpose.

Experimental Part

Mechanical Operation. The apparatus used in this work is that designed by Harkins and Ryan, and further developed by Harkins and Shadduck, and Harkins and Schuh. It has been described in detail in the publications of these workers. The only change made was that in the optical condensing system. The system of three lenses described by Schuh¹ was found to be unsatisfactory with the vertical carbon arcs at present in use. It was replaced by the single eleven centimeter aspheric condenser previously used.

A brief description of this apparatus follows. The cloud chamber is 6.3 centimeters in diameter by 1.5 centimeters deep. It is made of a well-annealed boro-silicate glass and has two small circular openings on one side. Into one of these the brass screw carrying the active deposit is fitted and through the other the gases examined are introduced. The floor of this chamber is the top of the piston covered with a layer of gelatine. The top of the chamber is an optically plane cover glass, coated on its under side with a thin layer of gelatine carrying a small amount of copper sulphate so as to make it a better electrical conductor. An electrical field of 250 volts between the top and bottom of

¹A. E. Schuh, University of Chicago Thesis.

the chamber, is used for sweeping out old tracks. The shutter for regulating the admission of the alpha particles into the chamber is in two parts. One, a small rectangular piece of heavy fibre board, is fastened to the piston. The other, a piece of the same material with a small rectangular opening in it, is fastened to the cover glass.

Expansions are produced in the chamber by means of the piston, which is fastened to a lever arm supported on a rotating cam. Two springs exert a tension on the lever arm and produce a sudden expansion when the support drops into the deep cut on the cam. The amount of expansion is regulated by an adjustable rubber stop, on to which the lever arm drops. The speed of the rotating cam determines the number of expansions per minute, which in general (in this work) was thirty-six. The cam is driven through a gear train by a direct current motor of one-eighth horse power.

The camera is synchronized with the expansion mechanism. It is a Universal No. 0 taking a 200 foot roll of 18 mm. by 35 mm. motion picture film. Eastman super-speed negative was used throughout. For each picture, two views of the chamber at right angles to each other are taken by means of a system of two plane and one 90° prism mirrors.

The light from two horizontal carbon arcs, drawing about 13 amperes at 180 volts, is passed through a condensing lens, a water cooling cell, and an adjustable slit on each side of the chamber.

A very good photograph of the apparatus is shown by Harkins and Schuh.¹

Preliminary Work with Nitrogen. The active deposit of thorium was used in all of the work described in this paper. This deposit furnishes alpha particles from thorium-C of 4.5 centimeters range to the extent of 35 per cent, and those from thorium-C' of 8.2 centimeters range to the extent of 65 per cent.

Photographs were taken of the tracks of about 29,000 of the long range particles in air, and of about 42,000 tracks in nitrogen but no disintegrations were observed. Considering the small number of tracks photographed, the absence of any disintegration is not unusual.

For the work with nitrogen it was thought that better conditions might result by an increase in the interval between expansions so that ten pictures per minute instead of thirty-six per minute would be obtained. Due to the resulting slow speed of the camera, however, and the consequent fact that at this slow speed the camera shutter had to be closed down to a very small opening which could not be adjusted accurately, the photographs obtained were not as good. These conditions could be remedied only by the use of a rather complicated and expensive system of gears in the gear train, and even then perfect synchronization between camera and

¹Harkins and Schuh, loc. cit.

expansion system might be difficult to obtain. Or alternatively, by a complete re-design and re-building of the apparatus. The apparatus was therefore restored to its original condition where thirty-six expansions per minute were made.

Work with Phosphine and Ammonia. Blackett¹ has taken pictures of the passage of alpha particles through hydrogen and has observed that all collisions in this gas are elastic (conservation of energy). It was to be expected therefore that in phosphine any disintegration of phosphorus would be inelastic, while all collisions with hydrogen would be elastic.

Due to the rapidity with which an expansion is produced in the cloud chamber (of the order of 0.001 sec.), it may be considered adiabatic, and the type of cloud produced dependent on the ratio of the initial and final temperatures which in turn is dependent on the initial and final volumes.² On this basis an expansion ratio giving a desired type of cloud for a diatomic gas could be used to calculate the necessary expansion ratio to produce a similar cloud in a monatomic, triatomic or other gas with a larger number of atoms. This ratio is given by the relation:

¹P.M.S. Blackett, Proc. Roy. Soc., A117, 124, (1927).
²C.T.R. Wilson, Proc. Roy. Soc., A87, 277 (1912).

$$\left(\frac{\theta_1}{\theta_2}\right) = \left(\frac{V_2}{V_1}\right)^{\gamma-1}$$

in which θ_1 and θ_2 are the initial and final temperatures corresponding to the volumes V_1 and V_2 when γ is the ratio of the specific heats of the gas considered. In a diatomic gas, nitrogen, the expansion ratio which gave the best tracks was 1.34. Taking the specific heats ratio of nitrogen as 1.37 and that of phosphine as 1.31, the corresponding expansion ratio for phosphine, using the above relation, would be 1.42. No values of the specific heats ratio could be found for phosphine (PH_3) so the values of ammonia (NH_3) as given in Mellor¹ were used.

The phosphine was prepared by the method of A. W. Hofmann² from phosphonium iodide. It was introduced into the chamber by the displacement of carbon dioxide. Using the expansion ratio calculated above, although a cloud was produced in the chamber, no tracks were made visible. Substituting a platinum tipped screw for the brass screw carrying the deposit, or sealing the source from the gas with mica caused no improvement. A decrease of the expansion or an increase to the limits of the apparatus likewise produced no tracks. An exactly similar phenomenon resulted when the

¹ J. W. Mellor, Comprehensive Treatise on Inorganic and
²Theoretical Chemistry, VIII (1928), p. 178.
Ibid., p. 807 and 825.

chamber was filled with ammonia (NH_3). In the latter case the failure was probably due to a solution of the ammonia in the water of the gelatine solution so that the apparent expansion ratio was a great deal less than the true ratio. A similar effect was probably the cause of the failure with phosphine. Since the maximum apparent ratio which could be obtained with our apparatus was insufficient to produce the tracks in these gases, this work had to be abandoned. When using these gases all metal in the chamber was protected against their action, and the gelatine which was used contained no copper sulphate.

Aluminium. For the first part of this work a thin sheet of aluminium, having a stopping power equivalent to 2.5 centimeters of air at 0°C . and 760 mm. pressure, was suspended in the chamber perpendicular to the cover glass, parallel to the face of the source screw and directly in the path of the rays. The stopping power of the sheet was calculated from its mass per unit area and on the basis that stopping power is proportional to the square root of the atomic number. Taking the range of the residual aluminium nucleus (recoil atom) after a disintegration (from the calculation of momenta distribution)¹ as 0.53 that of the incident alpha particle, and the remaining range of the effective alpha particles as about 6.0 cm., the range of the recoil atom would

¹E. Rutherford, Phil. Mag., 37, 569, (1919).

be 3.2 cm. Therefore in some cases the entire disintegrative process might be visible.

Photographs were taken of approximately 126,000 tracks of the 8.2 cm. range and 68,000 of the 4.5 cm. range. In no case was a disintegration observed.

Next, though it meant the sacrifice of the possibility of seeing the recoil atom, a sheet having a stopping power equivalent to 12 cm. was placed at a distance from the source equivalent to 1.6 cm. This caused the stopping of all alpha particles from the active deposit of thorium including those of abnormal range of 9.0 cm. and 10.9 cm. Therefore only H-particles would be visible on the far side of the sheet. Photographs were taken of approximately 301,000 tracks (thorium-C and C') and five H-particle tracks were observed. (A photograph of one of these tracks is shown in Figure 1). This is at the rate of seventeen H-particles per million incident alpha particles. Since, however, natural H-particles from the source might have a range in excess of 13.6 cm., the above figure is possibly too large. When a stopping power of 19.6 cm. (which would screen out abnormal range alpha particles of 14.2 and 17.4 cm. range*) was used, the yield was reduced to eleven H-particles per million, as obtained by observing photographs of 183,000 tracks.

*Alpha particles from the active thorium deposit of this range have been reported by Bates and Rogers, *Proc. Roy. Soc.*, A105, 97 (1924), but later workers were unable to check their results.

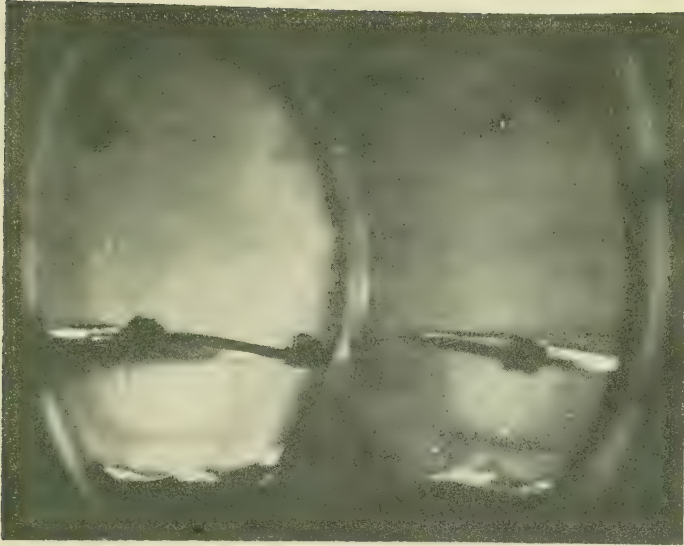


Figure 1

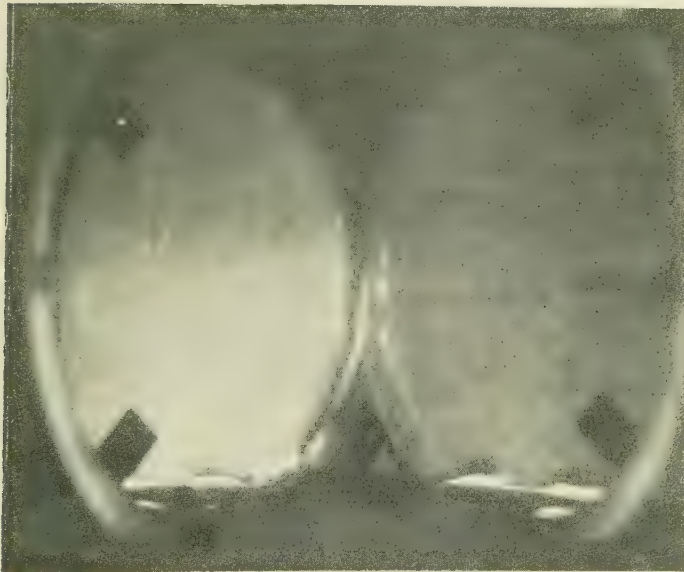


Figure 2

Since, however, the foils used were not degassed to remove any occluded hydrogen, it was not absolutely certain that all the H-particles observed were the result of an aluminium disintegration. Working with the particles of radium-C', Rutherford observed that the maximum range of H-particles from molecular hydrogen corresponded to 28 cm. air equivalent. It has also been observed that the range of H-particles is proportional to the cube of their velocity, that is, $V^3 = aR$, as is the case with alpha particles. On this basis, the maximum range of an H-particle from molecular hydrogen produced by an alpha particle of 8.2 cm. range is 34.6 cm. Therefore, if an aluminium sheet of this or a larger stopping power is used, only H-particles produced by a disintegration of aluminium will be observed on the far side of the sheet.

To obtain freedom from radioactive contamination in the chamber, the source was completely covered with the aluminium sheet. This removed the possibility of seeing any track but that of an H-particle in the chamber. The arrangement used was as follows: The sheet of stopping power equivalent to 35.9 cm. of air was sealed in the source holder at a distance of 0.3 cm. from the source. Both source and sheet were by this arrangement, outside of the effective part of the chamber.

As a result of this work, five H-particles were observed for approximately 548,000 alpha particles of thorium C and



Figure 3

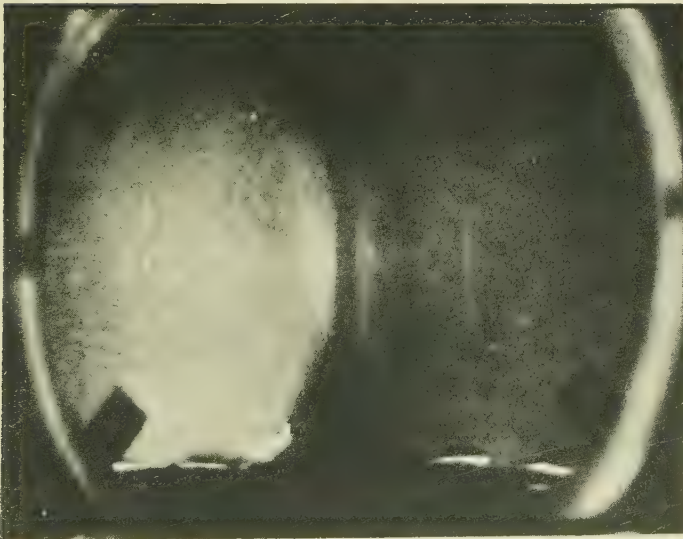


Figure 4

thorium C'. (Photographs of three of these tracks are shown in Figures 2, 3, and 4). This corresponds to a yield of 9 per million. On the basis of particles of range 8.2 cm., of which the number is approximately 356,000, the yield would be 14 H-particles per million thorium-C' alpha particles. In this series, using the heavy aluminium foils, the approximate number of alpha particles considered was determined by piercing the foil at the end of a run and taking pictures of about 100 expansions. (One of these photographs is shown in Figure 5).

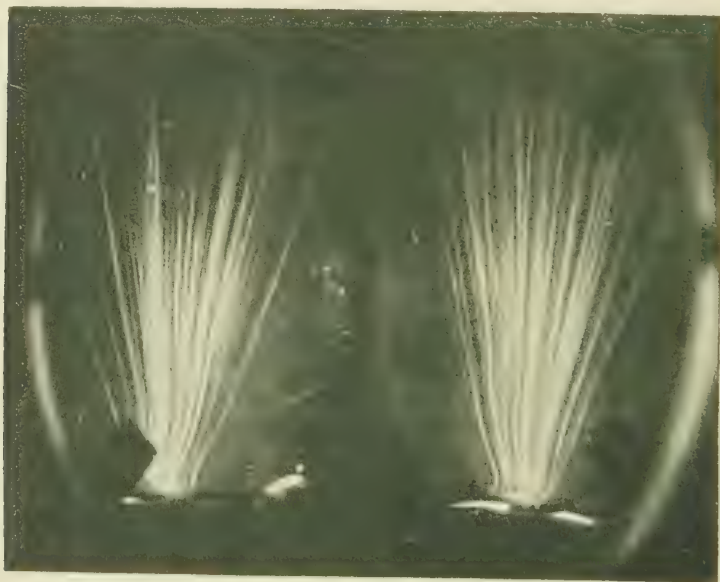


Figure 5.

Discussion of Results

Because of the possibility of H-particles having been produced from molecular hydrogen occluded in the aluminium foil, only the latter part of the work mentioned above can be considered as giving definite results. It is to be noted that the yield of nine protons per million incident alpha particles includes only those thrown off in the forward direction, and only a certain percentage of those. The work of Bothe and Franz¹ with a double Geiger counter and the "method of coincidence" indicates that at least two-thirds of the particles thrown off in the forward direction have been counted. The work of Rutherford and Chadwick indicates that the yield in the backward direction from aluminium is approximately 45 per cent of that in the forward direction. Considering that both these values apply here, the total yield from aluminium would be approximately 20 protons per million incident alpha particles.

It is rather difficult to compare this value with values obtained by other workers with aluminium. In their early work Rutherford and Chadwick² give a yield from aluminium of 2 protons per million incident alpha particles of radium

¹W. Bothe and H. Franz, loc. cit.

²E. Rutherford and J. Chadwick, Phil. Mag., 42, 824 (1921).

active deposit. They do not indicate whether this yield represents that from the entire solid sphere or not. In the same article for a rough comparison of yields they give a yield of 0.7 (protons per minute per mgm. of radium) from nitrogen and 1.1 from aluminium. Later, Chadwick¹ speaks of the yield from nitrogen as being 20 protons per million. If the early relation between the yields from the two elements is accurate, their yield from aluminium would be 31 protons per million. Others, however, referring to their work seem to consider their yield to be 2 protons per million from aluminium. Schmidt,² at Vienna, obtains a yield of approximately 30 protons per million from aluminium, presumably a total yield. This figure is for 1 cm. equivalent absorption.

Holoubek,³ at Vienna, is the only one who has done work with aluminium in a Wilson chamber. He, however, bases all his conclusions on visual observation and not as was done here on a photographic record. There is another point in his work which is of prime importance. It is the number of expansions per minute which he uses.

In the work at Chicago great pains have been used to prevent radioactive contamination in the chamber. But, using

¹J. Chadwick, Proc. Roy. Soc., A123, 383 (1929).

²E. A. W. Schmidt, Z. Physik., 42, 721 (1927).

³R. Holoubek, Z. Physik, 42, 704, (1927).

all conceivable precautions, contamination was still present. In the first examination of one reel of film representing 165,000 alpha particles, nine tracks were obtained, which came from the direction of the source. A statistical consideration of all the stray tracks in the chamber and the point from which they originated indicated that seven stray tracks could be expected from the source. A close examination of the original nine showed that seven of them were broad and diffuse, while two only were sharp and faint. Due to the high speed of H-particles, they leave a sharp faint track in the cloud. The appearance of these tracks is quite different from those of alpha tracks. This can be seen by comparison of Figure 5 with one of the other pictures. It was also distinctly noticed by the workers with nitrogen. So it is safe to assume that in this case two proton tracks were obtained.

Our work showed that in some cases the stray cloud track was not swept out before a new expansion started so that the same track was photographed in two successive pictures. This was at a speed of thirty-six expansions per minute. In no case of an observed disintegration did a track appear on the photograph just before or just after.

In Holoubek's work he refers to "contamination" and he was presumably bothered by it. He also specifically states

that yield is dependent on expansion rate,¹ and uses expansion speeds of from 60 to 130 per minute. If tracks are not always removed at speeds of 36 expansions per minute, certainly they would be less likely to be at 104 expansions per minute. It is illogical that the number of protons produced should depend on the method of observing them. Also, by visual observation it becomes somewhat difficult to distinguish an H-particle track from the thick track of an alpha particle, particularly when the view of it lasts less than half of a second. So that while it is possible that H-particles from aluminium were obtained in Holoubek's work, the yield is by no means as high as that given.

The early work of Chadwick² indicates that it is doubtful if particles of range under 4.5 cm. are capable of disintegrating aluminium. His later work³ indicates that the yield for particles of less than this range is very small, and that no disintegration at all occurs for particles of range less than about 3.5 cm.

Holoubek uses the alpha particles of radium-F, which have a range of 3.7 cm. As mentioned above, the particles used in this work have ranges of 8.2 cm. and 4.5 cm. and should therefore give higher yields than were obtained by him. His yield is about 30 per million whereas the yield in this work is no greater than 20 per million.

¹R. Holoubek, Z. Physik, 42, 712 (1927).

²J. Chadwick, Phil. Mag. 42, 825 (1921).

³J. Chadwick, Proc. Roy. Soc., A123, 384 (1929).

In the same work Holoubek offers evidence for the disintegration of beryllium, carbon, oxygen, and iron, which agrees with the results obtained by Kirsch and Pettersson, who used the scintillation method, but is entirely at variance with the results of Rutherford and Chadwick, and Bothe and Franz. The results of our work indicate that his results are very doubtful.

Summary

Figure 1, as mentioned above, shows an H-particle track from aluminium, which has a stopping power of 12 centimeters. It can not be definitely stated that this particle is produced by an aluminium disintegration, since natural H-particles from the source were not screened out.

Figures 2, 3, and 4 show H-particle tracks produced by the disintegration of aluminium and having ranges in excess of about 36 centimeters.

Figure 5 shows alpha particle tracks from the active thorium deposit.

A method of observing an atomic disintegration of aluminium is described. This method has the possibility of being used to obtain complete dynamical pictures of the disintegrative process, similar to those obtained in nitrogen.

The yield of H-particles per million incident alpha particles from the thorium active deposit was observed as nine for ranges in excess of about 36 cm. air equivalent.

The writer gratefully acknowledges the kind direction and assistance of Professor William D. Harkins in this work.

